# NbSe<sub>3</sub>: Effect of Uniaxial Stress on the Threshold Field and Fermiology

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We have measured the effect of elastic strain  $\epsilon$  on the threshold field  $E_T$  for the motion of the higher temperature charge density wave(CDW) in NbSe<sub>3</sub>. We find that  $E_T$  exhibits a critical behavior,  $E_T \sim (1 - \epsilon/\epsilon_c)^{\gamma}$  where  $\epsilon_c$  is about 2.6%,  $\gamma \sim 1.2$ . This expression remains valid over more than two decades of  $E_T$ , up to the highest fields of about 1.5 kV/m measured using pulse techniques. Neither  $\gamma$  nor  $\epsilon_c$  is very sensitive to the impurity content of the sample. The transition temperature is linear with  $\epsilon$ , and  $dT_P/d\epsilon = 10$  K/% shows no anomaly near  $\epsilon_c$ . The slope of the narrow band noise frequency vs. the CDW current does not change appreciably with  $\epsilon$ . Shubnikov-de Haas measurements show that the extremal area of the Fermi surface decreases with increasing  $\epsilon$ . We conclude that there is a very intimate relationship between pinning and the Fermiology in NbSe<sub>3</sub>.

#### Introduction:

The recent discovery of the Aharonov-Bohm effect exhibited by the sliding CDW in NbSe<sub>3</sub> has revived interest for the field of CDW<sup>1</sup>. Non-linear conductivity is the outstanding characteristic of charge density wave materials<sup>2</sup>. The presence of a threshold field  $E_T$ , above which the resistance decreases, is the signature that the CDW can be made to move under a small electric field $^{3,4}$ . The dependence of  $E_T$  on temperature (T), number of impurities  $(n_i)^{5,6}$ , contact position, size<sup>7</sup>, pressure<sup>8</sup> and uniaxial stress<sup>9,10</sup>, has been extensively reported. Here we report further studies on the effect of elastic, uniaxial stress  $\sigma$  on  $E_T$  for the upper CDW in NbSe<sub>3</sub>. This paper will show that  $E_T \sim 1/|\sigma - \sigma_C|$ , where  $\sigma_C \approx 260$ GPa and that this is related to the change in Fermiology as shown from low temperature Subnikov-de Haas (SdH) studies.

# I. EXPERIMENTAL TECHNIQUES AND RESULTS

## A. Samples

The experiments were conducted on nominally pure as well as Fe doped NbSe<sub>3</sub> samples. Fe doping was achieved by mixing either 4.7% or 7% of Fe in the starting materials; about a tenth that much doping is expected in the resulting whiskers<sup>11</sup>. Samples of medium purity with a RRR of 70 were grown in house; the high purity samples, RRR >200, were provided by R.E. Thorne. The samples were mounted on a stressing device described elsewhere<sup>12</sup>. Uniaxial stress was applied along the needle axis, the **b** crystal axis. The strain  $\varepsilon$  was directly measured, and can be converted to stress using the Young's modulus , S<sub>22</sub>  $\approx$  100 GPa<sup>10</sup>. Four electrical contacts were made using conducting silver paint. Epoxy overlaid these contacts and formed the mechanical grips. Typical sample dimensions were 2,000x 20 x1  $\mu$ m<sup>3</sup>.

#### B. Effect of $\varepsilon$ on the Upper CDW

At low strain and low fields, the threshold field was determined using the conventional lockin or dV/dI technique. At high  $\varepsilon$ , where high electric fields are required to reach  $E_T$ , the pulse method was used. The duty cycle was less than 1%; with typical pulse width of 10  $\mu$ s, and period 1 ms. The pulsed current and voltage were measured using a two channel boxcar signal averager, EG&G model 162. In this case,  $E_T$  was estimated from the plot of the chordal resistance R vs E or from the numerical derivative  $\Delta V/\Delta I$ . Previous studies<sup>9,10</sup> have shown that uniaxial stress affects  $E_T$  indirectly by  $\varepsilon$  induced changes in  $T_P$  and directly by enhancing the pinning strength. It was shown that the indirect effect can be disentangled by conducting the experiments at a constant reduced temperature  $t = T/T_n(\varepsilon)$  where  $T_n(\varepsilon)$  is defined at the peak in  $dR(\varepsilon)/dT$ . Constant t=0.70 was achieved by adjusting T for each value of  $\varepsilon$ . This value of t corresponds to the minimum value  $E_{min}$  on the  $E_T$  vs. t curve. It was previously shown that  $E_{min}$  is proportional to the impurity concentration<sup>6,7</sup> and assumed to correspond to bulk pinning rather than contact pinning. Although this paper is devoted to the study of the effect of  $\varepsilon$  on  $E_{min}$ , for the sake of simplicity we will refer to it as the threshold field, or simply  $E_T$ .

FIG. 1.  $E_T$  vs  $\varepsilon$  for a nominally pure sample. The inset shows a semilog plot of the same data.

Figure 1 shows a typical plot of  $E_T$  vs.  $\varepsilon$  for an arbitrarily selected sample.  $E_T$  increases weakly at low strain and diverges near  $\varepsilon_c=2.6\pm0.3\%$ . The semi-log plot of the same data shown in the inset indicates that  $E_T$  increases faster than a single exponential. In the few cases where we were able to pull beyond 2.6% strain,  $E_T$  exhibited a peak, decreasing above 2.6  $\pm$  0.3%. Figure 2 shows the strain dependence of  $T_P$ .  $T_P$  decreases linearly with increasing  $\varepsilon$  up to 3 % at a rate  $dT_P/d\varepsilon=10~{\rm K/\%}$ . There is no apparent feature around  $\varepsilon=2.6$ %, where  $E_T$  diverges. The inset in Figure 2 shows a plot of R vs T for

different values of  $\varepsilon$ . Note that the resistance anomaly  $\Delta R(\varepsilon) = R_p(\varepsilon) - R_{fit}(\varepsilon)$  is independent of  $\varepsilon$ , where  $R_p(\varepsilon)$  corresponds to the peak resistance for a given  $R(\varepsilon)$  vs T plot, and  $R(\varepsilon)_{fit}$  is the linearly extrapolated resistance at T peak from above 150 K. This result suggests that the CDW conductance ( $G_{cdw} \sim n_{cdw} e \mu$  where e is the charge of the electron and  $\mu$  the cdw mobility) at very large electric fields is independent of  $\varepsilon$ , which in turn implies that the fraction of condensed electrons  $n_{cdw}$  does not change appreciably with  $\varepsilon$ . This is consistent with narrow band noise measurements which showed almost no change in the slope of the CDW current vs the narrow band frequency  $(dI_{cdw}/dF)$  with  $\varepsilon^{13}$ .

FIG. 2. The CDW transition temperature  $T_{p1}$  vs. strain.  $T_{p1}$  decreases linearly with  $\varepsilon$  up to  $\varepsilon = 3$  %. Typical  $R(\varepsilon)$  vs. T plots are shown in the inset.

#### C. Effect of $\varepsilon$ on the Fermi Surface

This section looked for a connection between the effect of  $\varepsilon$  on the Fermiology and the results reported in the previous section. The effect of  $\varepsilon$  on the dominant frequency of the Shubnikov-de Haas oscillations is reported. The magnetic field  $B_a$  was parallel to the  $(\mathbf{b}, \mathbf{c})$  plane and perpendicular to the smallest extremal area of the Fermi surface, with typical frequency of 0.28 MG at  $\varepsilon = 0^{13,14}$ . Two methods were used. The first method is the conventional method,  $B_a$  was increased slowly with the sample under constant strain. In the second method,  $B_a$  was constant while sweeping  $\varepsilon$ . The experiments were conducted at constant T between 3.0 and 4.2 K.

Figure 3a shows a typical plot of R vs H obtained using the conventional method, the inset shows dR/d(1/H) vs 1/H. The extremal area A was estimated from a plot of n vs 1/H for each value of  $\varepsilon$ . Figure 3b shows that A decreases nearly linearly with increasing  $\varepsilon$ . A detailed study of the effect of uniaxial stress on the Fermi surface will be reported elsewhere. In this paper we note that uniaxial stress suppresses A linearly at the rate of 0.09 MG/%, suggesting that the whole pocket would be wiped out for  $\varepsilon \leq 3\%$ . A study of the strain dependence of the conductance at low temperature shows that 90% of the conductance is wiped out for  $\varepsilon \approx 2.6\%$ . This suggests that this pocket plays a predominant role in the normal state conductance of NbSe<sub>3</sub> at low T.

FIG. 3. Fig 3a shows a typical plot of R vs H which exhibits a the Shubnikov-de-Haas oscillations. The derivative dR/d(1/H) vs 1/H is shown in the inset. In fig3b the extremal area, in units of kG, decreases smoothly with increasing  $\varepsilon$ .

The second method is equivalent to fixing the Landau tubes and shrinking the Fermi surface through them under the influence of  $\varepsilon$ . This leads to oscillations in the R vs  $\varepsilon$  plots as shown in Fig. 4a. A systematic study of R vs  $\varepsilon$  for different values of  $B_a$  allows us to follow

the strain and the field at which a given Landau tube is crossed. The results are summarized in Fig. 4b., which shows a plot of  $\varepsilon$  vs  $B_a$  for each Landau tube identified by the integer next to its curve. The trajectory of a given Landau tube is nearly linear. This is consistent with the linear relationship between A and  $\varepsilon$  observed using the conventional technique. The solid lines in the figure are a guide to the eye. Note that at  $B_a=0$  T, all the lines converge to nearly the same  $\varepsilon_c^H\simeq 2.6$  %. This suggests that this piece of the Fermi surface would be wiped out at about 2.6 %. Below we will also show that  $\varepsilon_c^H$  is equal to the critical strain  $\varepsilon_c^{ET}$  derived from the critical plot of  $E_t$  of the upper CDW.

FIG. 4. Fig 4a shows the oscillatory R vs  $\varepsilon$  plots for B= 5.4 Tesla. The oscillations are attributed to the intersection of the Landau tubes with the shrinking Fermi surface. Figure 4b is a representation of this intersection in the  $(\varepsilon, B)$  space. The integers in the box correspond to the indices of the Landau levels.

#### II. DISCUSSION

Possible pinning mechanisms of the CDW are: bulk impurity pinning as discussed by Fukuyama-Lee-Rice<sup>16</sup> (either strong or weak), commensurability pinning by the underlying lattice, or pinning by other defects such as surfaces, dislocations or contacts. The results in Fig. 3 could be due to one or a complex combination of the following effects: (1) strain induced enhancement of the weak impurity pinning potential; (2) strain induced crossover from weak pinning to strong pinning; (3) strain induced incommensurate to commensurate transition; or (4) strain induced enhancement of contact pinning. We now discuss each one of these effects separately, in inverse order of their likelihood.

Stress induced enhancement of contact pinning is very unlikely. If this were the case, one would also expect a similar stress induced enhancement of  $E_T$  for the lower CDW. However, previous studies have shown that stress does not enhance  $E_T$  for the lower CDW<sup>9,10</sup>. In addition, Y. Tseng et al. have shown<sup>9</sup> in the case of the upper CDW,  $E_T$  can be separated into two components, one attributed to contact pinning, and the other to bulk impurity pinning. They have argued that uniaxial stress does not enhance contact pinning. It also seems unlikely that uniaxial stress can affect surface pinning to that extent; if it did, our thinner samples would have shown a stronger effect.

The FLR model considers two possible kinds of impurity pinning: strong pinning and weak pinning. Several experiments indicate that pinning in NbSe<sub>3</sub> is due to weak pinning<sup>6,7</sup>. Stress-induced crossover from weak pinning to strong pinning could be considered, in which case one would expect to see a change in the exponent  $\gamma$  with  $\varepsilon$ .  $\gamma$  is defined in the next paragraph. The results show that  $\gamma$  is independent of  $\varepsilon$  and rule out this possi-

bility as well. This is also supported by the fact that the same  $\gamma$  is obtained for the samples doped with Fe, which may be considered as a strong pinning impurity.

FIG. 5. A critical plot of  $E_T$  for five different samples. The full triangles and the crossed squares correspond to Fe doped samples, 0.7% and 0.47% respectively. The normalized threshold  $e_T = E_T(\varepsilon)/E_T(0)$  is shown in the inset. Note that all five set of data fall on the same line.

## Experimental

search for an incommensurate-commensurate transition (ICT) in CDW systems has not provided any clear evidence for these effects<sup>17</sup>. A stress-induced ICT would lead to changes in  $dT_P/d\sigma$  as well as soliton-like behavior near commensurability. Figure 2 shows that  $T_P$  does not show an anomaly near  $\varepsilon_c$ . Further, according to Fisher and Fisher<sup>18</sup> the approach to commensurability should behave critically with an exponent of  $\frac{1}{2}$  (for 2-D) or be logarithmic. Figure 5 shows such a critical plot of  $E_T$  vs  $(1 - \varepsilon/\varepsilon_c)$  in a log-log scale. A plot for the normalized threshold field

$$e_T = E_T / E_{T_0} = (1 - \varepsilon / \varepsilon_c)^{\gamma} \tag{1}$$

where  $E_{T_0}$  is the threshold field at zero strain , and  $\varepsilon_c$  and  $\gamma$  are adjustable parameters is shown in the inset. In the following we will replace  $\varepsilon_c with \ \varepsilon_c^{E_T}$  in order to differentiate it with the critical strain defined from the fermiology study. Note that the results for five different samples with different impurity content, and Fe impurity fall along the same line, with nearly the same parameters. A list of the values of  $\varepsilon_c^{E_T}$  for different samples is shown in Table 1. Although the figure is in qualitative agreement with Fisher and Fishers prediction that  $E_T$  should behave critically in an ICT, the exponents are not in quantitative agreement with the model. Our larger samples are most likely 3-D, and the exponent is not 1/2; therefore the critical behavior cannot be explained by a simple approach to commensurability. On the other hand, an argument in favor of ICT can be made based on the divergence of  $E_T$ . Since commensurability pinning is much stronger than impurity pinning,  $E_T$  is much more sensitive to stressinduced ICT than  $T_P$  is. This issue could be resolved using structural studies as a function of strain.

**Table 1:** The fitting parameters  $\varepsilon_c^{E_T}$  and  $\gamma$  are shown together with other relevant parameters such as the nominal purity, the RRR, and the threshold field  $E_T$ . Only the nominal Fe doping levels are given.

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		$\varepsilon_c^{E_T}\%$		$E_T (V/m)$
Pure	250	2.6	1.58	
Pure	200	3	1.23	
Pure	-	2.6	1.23	
Pure	-	2.6	1.66	
4.7% Fe	-	3.2	1.14	
4.7 %  Fe	-	2.6	1.08	
7% Fe	-	2.7	1.23	63

According to the FLR theory<sup>16</sup>, the threshold field for weak pinning is given by:

$$eE_T \lambda \propto \frac{\Delta^2}{E_F} (\xi_x, \xi_y, \xi_z n_i^2) V_0^{(\frac{4}{4-D})}$$
 (2)

where  $\lambda$  is the wavelength of the CDW,  $\xi_x, \xi_y$ , and  $\xi_z$ are coherence lengths for the CDW amplitude,  $E_T$ the threshold field, e the electric charge,  $E_F$  the Fermi energy,  $V_0$  the impurity potential and D the dimensionality<sup>16</sup>. Stress could effect any or all of the parameters in Eq. 2. However, for the sake of simplicity we will discuss separately the terms that are susceptible to change with  $T_P$ . In the conventional BCS model the CDW gap is proportional to  $T_P$ ,  $\Delta/T_P = 4.8$  for NbSe<sub>3</sub><sup>1</sup>. As in previous pressure work<sup>8</sup>, uniaxial stress induced enhacement of the electron-phonon coupling constant could be considered. However, it would take more than an order of magnitude of change in order to account for our results. On the other hand if, in a first approximation, one assumes that this ratio is not affected by  $\varepsilon$ ,  $\Delta$  would decrease with  $T_P$  which would lead to a decrease in  $E_T$ , contrary to our results. Another possibility is a straininduced decrease in  $E_F$ . But the normal state conductivity around  $T_P$  is a weak function of  $\varepsilon$ , even up to 3%, suggesting that  $E_F$  is not strongly affected by  $\varepsilon$ . One likely possibility is that  $V_0$  is strongly affected by stress. Suppose there is a stress-induced tuning of the matching between the phase and wavelength of the CDW and the Friedel oscillations<sup>19</sup>. Then, although the changes in  $E_F$  due to the vanishing of this small pocket could be negligible, it could be sufficient to lead to a rapid increase of  $V_0$ . This mechanism would be independent of the type of impurity and concentration, in agreement with our experiments. One other possibility is that the pocket screens the impurity, and  $V_o$  increases when the pockets disappears. Below we will discuss the difference between the upper and lower CDW.

In a study of the combined effect of magnetic field and strain, Parilla et al.<sup>20</sup> have shown that uniaxial stress and  $\mu_0 H$  act on the same piece of the Fermi surface. This was confirmed by Y.T. Tseng et al.<sup>21</sup> who observed pronounced effect of strain on the resistance and thermopower of NbSe<sub>3</sub> below 59 K. Jianhui et al.<sup>22</sup> have conducted NMR experiments to study the density of states on the different chains in NbSe<sub>3</sub>. Although magnetic fields effects on the ohmic regime, below  $E_T$ , are much more pronounced below the second transition than below  $T_{p1}$ , their results show that most of the changes in FS is due to changes in density of states on the chain associated with the upper CDW rather than the lower CDW. This supports the notion that the strain-induced changes in  $E_T$  of the upper CDW are associated with changes in the Fermi surface most closely associated with the chain corresponding to the upper CDW. The relatively small effects on the density of states associated with the lower CDW could account for the rather weak effect on the  $E_T$ of the lower CDW.

#### III. CONCLUSION

It was previously reported that uniaxial stress enhances  $E_T$  for the upper CDW in NbSe<sub>3</sub>. In this paper we report a systematic study of the effect of  $\varepsilon$  on  $E_T$ ,  $T_P$ , and the Fermi surface of this compound. We show that the divergence of  $E_T$  near 2.6% strain is intimately related to stress induced changes in fermiology. We propose that the two most likely possibilities for this phenomena are (1) a stress induced incommensurate to commensurate transition (2) or more likely an  $\varepsilon$  driven matching of the Friedel oscillations and the CDW oscillations of the upper CDW. Structural studies under stress should give a further insight into the subject.

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